

**U.S. DEPARTMENT OF ENERGY  
NUCLEAR ENERGY RESEARCH INITIATIVE  
ABSTRACT**

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**PI:** David Bartels

**Proposal No.:** 99-0276

**Institution:** Argonne National Laboratory

**Collaborators:** Atomic Energy of Canada LTD-Chalk River Laboratories

**Title:** Radiation-Induced Chemistry in High Temperature and Pressure Water and Its Role in Corrosion

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Commercial nuclear reactors provide a source of heat, used to drive a "heat engine" (turbine) to create electricity. A fundamental result of Thermodynamics shows that the higher the temperature at which any heat engine is operated, the greater its efficiency. Consequently, one obvious way to increase the operating efficiency and profitability for future nuclear power plants is to heat the water of the primary cooling loop to higher temperatures. Current pressurized water reactors run at roughly 300° C and 100 atmospheres pressure. Designs under consideration would operate at 450° C and 250 atmospheres, i.e. well beyond the critical point of water. This would improve the thermodynamic efficiency by about 30%. A major unanswered question which we propose to address, is what changes occur in the radiation-induced chemistry in water as the temperature and pressure are raised beyond the critical point, and what does this imply for the limiting corrosion processes in the materials of the primary cooling loop?

The cooling water of any water-cooled reactor undergoes radiolytic decomposition, induced by gamma, fast-electron and neutron radiation in the reactor cores. Unless mitigating steps are taken, oxidizing species produced by the coolant radiolysis can promote intergranular stress-corrosion cracking and irradiation-assisted stress-corrosion cracking of iron- and nickel-based alloys, and generally altered corrosion rates of iron- and nickel-based alloys, and zirconium alloys in reactors. One commonly used remedial measure to limit corrosion by oxidizing species is to add hydrogen in sufficient quantity to chemically reduce transient radiolytic primary oxidizing species (OH, H<sub>2</sub>O<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>), thereby stopping the formation of oxidizing products (H<sub>2</sub>O<sub>2</sub> and O<sub>2</sub>). It is unclear whether this will be effective at the higher temperatures proposed for future reactors.

The unique collaboration proposed here is ideally suited to discover much of the necessary fundamental information. Electron pulse radiolysis coupled with transient absorption spectroscopy is the method of choice for measuring kinetics of radiation-induced species, and also yields for fast electron and gamma radiation. Workers at AECL have experience in making radiation yield and kinetics measurements to temperatures as high as 300°C, using a 2.5 MeV electron accelerator at Chalk River Laboratories. There is extensive experience at AECL in modeling of radiation physics and radiation chemistry in nuclear reactors. To reach higher pressures, greater penetrating power of the electron beam is desired, and shorter pulses are required to measure faster chemical transients at higher temperature. The Argonne Chemistry Division linac is capable of producing 20 MeV

electron pulses of 30 picoseconds duration, and the principal investigators at Argonne have extensive experience in measuring transients on a nanosecond and sub-nanosecond timescale. This capability will make possible the measurement of radiation yields and kinetics in water under conditions that have never been previously investigated.

The subject of this proposal touches on several areas of research mentioned in the NERI call for proposals. At its heart, the work is fundamental chemical science which can be applied to both current and future reactor problems, and other areas of endeavor such as supercritical water oxidation technology. The direct application to nuclear engineering research is the design of reactors with higher performance and efficiency.